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Photoluminescense of isolated quantum dots in metastable InAs arrays

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Abstract. Certain cases are studied, in which quantum dots showing no collective characteristics appear. It is demonstrated that isolated quantum dots are typical of the metastable arrays. The light radiated by these nanoislands has a narrower spectral band as compared to other dot arrays. This effect should be taken into account for the development of quantum dot lasers, in particular, for vertically cavity surface emitting lasers.

Introduction

It has been observed that in metastable arrays the quantum dot (QD) 0D-properties are more prominent than in case of stable QDs, which have square bases and are organized into the dense 2D square lattice [1]. This phenomenon is not explained by the available calculations [2] justifying the metastable QD existence. The narrow photoluminescence (PL) band of metastable QDs provides an advantage for device application. But structural (morphological) instability and tendency to coalescence are restrictive factors for the metastable array application, therefore they need thorough studies. Up to now, the questions remain: what are the metastability consequences on the QD array and which mechanisms are responsible for its PL spectrum?

1. Samples

The structures comprised arrays having 1, 6, 10 or 15 InAs QD layers in GaAs matrix confined on both sides by $Al_{0.25}Ga_{0.75}As/GaAs$ superlattices. Singular and vicinal GaAs(100) substrates misoriented towards [001] and [010] directions with 3°, 5°, 7° angles were used. For the single-layer structures (2D arrays) the submonolayer migration enhanced epitaxy (SMEE) mode was applied. For the multi-layer structures (multiply stacked arrays) the combined MBE + SMEE technique was used. The adjacent InAs layers were separated by the GaAs spacer. In different samples it had 8, 10 or 12 nm thickness. Each SMEE InAs layer was produced by the repeated deposition cycle including 0.5 monolayer (ML). In followed by the surface exposure to As_4 flux during $\tau = 10$ s. The InAs QD layer growth temperature was 470°C, growth rate – 0.1 ML/s. QDs formed by the deposition of 1.8, 2.5 and 3.0 InAs ML were studied.

2. Isolated InAs QDs in 2D arrays on the GaAs vicinal surface

STM images reveal the morphological metastability of resulting QD arrays [3]. On the singular GaAs surface this growth mode forms the well-ordered InAs QD array with a

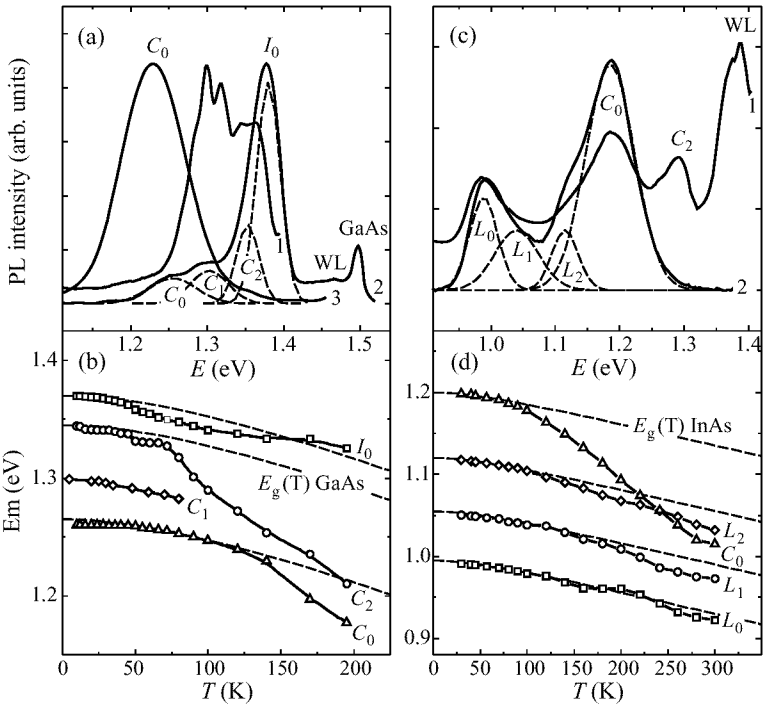

Fig. 1. PL spectra at $T = 5$ K (a,c) and PL peak energy *vs* temperature (b,d) for the 2D arrays of 1.8 ML InAs QD (a,b) and for the 10 multiply stacked arrays of 2.5 ML InAs QD (c,d) on GaAs (100) substrates: (a, curve 3), (c) and (d) — singular substrate; in other cases — 7°[001] vicinal substrate. Selective excitation: (a, c, curves 1) — WL excitation, in other cases — GaAs matrix excitation. Dashed lines in (a), (c) — deconvolution of PL spectrum (curves 2).

chain-like structure organized along the [001] direction. For the 1.8 ML InAs the linear density of QDs in the chain is 0.1 nm^{-1} and the area coverage is 0.3. The QDs have a rectangular 5 × 12 nm base. The vicinal surface is characterized by a strong step bunching effect. For the 7° angle the average statistical terrace size is 50 nm. The QD arrays are located on the terraces. The chain-like structure remains.

The PL studies have shown that samples with a small substrate misorientation (including singular) have one QD group radiating in the ground and excited states: C_0 and C_n bands ($n = 1, 2$ and more, depending on the excitation conditions). Full width at half maximum (FWHM) for the C_0 band is: 95 meV for a singular substrate and 80 meV for the 3° misoriented one. In the samples with a high misorientation degree (5°, 7°) the second QD group appears, which gives a more intensive I_0 band with FWHM = 35 meV in the short-wave part of the PL spectrum (Fig. 1a).

The PL spectrum *vs* the excitation energy, as well as the PL peak energy and FWHM *vs* temperature showed that these QD groups (CQD and IQD) had basically different properties.

Selective excitation showed that CQD group is connected to the wetting layer (WL), and IQD – only to the GaAs matrix (Fig. 1a). The PL excitation spectra demonstrated the absence of carrier exchange between CQD and IQD groups.

The CQD PL peak energy and FWHM *vs* temperature is the evidence of carrier redistribution from small to large QDs (Fig. 1b). As the temperature grows, the C-bands

get narrower and reveal a red shift considerably exceeding the temperature shifts of the InAs and GaAs band gaps. Presence of such temperature redistribution indicates a close vicinity of CQD (15 nm from the tunnelling length calculations). Such QDs constitute the chain-like structure (CQD — conjunct QDs).

The I_0 band evolution at increasing temperature shows the absence of carrier exchange between the IQD. Their FWHM increases and the red shift follows the GaAs band gap change (Fig. 1b). Consequently these QDs are separated not only from the CQD group but also from each other (IQD — isolated QDs). This can be caused by the WL local absence due to its ruptures at step edges for samples with a high degree of step bunching [4].

The metastable array formation can probably be caused by the combination of SMEE mode ($\tau = 10$ s), which efficiently increases the adatom migration length, and the low growth temperature, which decreases the adatom mobility on the surface. By this the conditions for the appearance of GaAs substrate elastic anisotropy are established, which is expressed as the distinct direction [001], along which QD chains are arranged. It has been previously noted [3] that at the alternate submonolayer deposition the chain formation is accompanied by the generation of WL corrugations. It can be assumed that the overlap of corrugation structures and the system of multiatomic steps can result in WL ruptures on the vicinal surface. In this way the elastic strain relaxation mechanism, which competes with QD formation, is eliminated and isolated QDs are formed.

3. Isolated QDs in multiply stacked arrays

TEM images of multiply stacked arrays [5] demonstrated large QDs (LQD) along with CQD in the upper layers. LQD had lateral sizes ranging within 20–40 nm.

PL spectrum for single-layer structures had a previously-mentioned C_0 band with a 1.2 eV peak energy and 95 meV FWHM ($T = 5$ K). In the structures with six QD layers and 8 nm thick spacer the main C_0 band developed a long-wave shoulder. When ten InAs layers were grown the main C_0 maximum was accompanied by the long-wave maximum L_0 having the 0.98 eV energy (Fig. 1c). The multiplication of 15 layers made the L_0 maximum domineer in the PL spectrum in the 0.96 eV position (FWHM = 50 meV). Under room temperature its position was at 1.4 μm.

The increase of spacer size inhibited the new L_0 band formation in the PL spectrum. It was enough to increase the spacer thickness to 12 nm to make it disappear.

In our opinion a new L_0 band appearance was caused by the formation of islands with sizes larger than those of CQD in the InAs layers. The gap between C_0 and L_0 bands reached 200 meV. A new LQD group could appear as the result of QD coalescence in the InAs upper layers [5, 6]. The SMEE method promotes the growth of metastable QDs, which coalesce readily, and it stimulates the QD coalescence. Large InAs islands appear in the upper layers, they have the lateral sizes about 30 nm and produce L_0 radiation at 1.3 μm.

The deconvolution of multiply stacked QD PL spectrum into gaussians gives 4 components (Fig. 1c), the exterior ones are the L_0 and C_0 bands, they result from the transitions between the ground states. The L_1 and L_2 transitions take place with the involvement of large QD excited states.

The studies of PL spectrum dependence on the excitation energy (Fig. 1c), as well as the PL peak energy on temperature (Fig. 1d) enabled to draw the following conclusions.

Large QDs (L_0, L_1, L_2 bands) lie on the WL, but are not connected to each other. There is no carrier redistribution within the LQD group in the whole temperature interval. Consequently the carrier exchange between LQD is either missing or impeded. Because

of this such QDs are also isolated [7]. Such isolation can be caused by the low density in the coalesced QD group. The dot–dot separation exceeds the carrier diffusion length in the WL, taking into account the carrier dissipation across the shallow CQD quantum wells.

The InAs layer multiplication on the vicinal GaAs(100) substrates results in the I_0 band (physically isolated QDs) disappearance from the PL spectrum. We suppose that IQD serve as the elastic strain relaxation mechanism in the bottom layer, therefore they do not reproduce in the upper layers.

In this way, due to the lack of carrier exchange the isolated QDs in IQD and LQD groups demonstrate only individual properties. The PL bands corresponding to them are two times narrower than the conjunct QD bands. The inhomogeneous broadening of IQD depends on the size variation only for the narrow terraces, on the edges of which the WL rupture takes place [4]. FWHM of LQD is just a part of the inhomogeneous CQD broadening, as only the largest CQD take part in the LQD group formation.

Conclusion

QD metastability causes the array separation into two subsystems. On the vicinal surface the separation results from the morphological instability (CQD and IQD); on the stack upper layers it is caused by the tendency to coalesce (CQD and LQD). Therefore we can conclude that isolated QDs accompany metastable QD arrays. The specific features of this phenomenon and the inevitable development of the isolated QD subsystem are discussed.

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